NSLS-II Beamline Development Proposal

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Coherent Diffraction Imaging (CDI)

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A. Science Case

- 3D Imaging of crystal shapes and strain fields inside crystals on the nanometer scale
- Evolution of shape/strain under working conditions or manipulation/deformation/indentation
- Ptychographic imaging for extended objects and biological samples using phase contrast
- Cryogenic sample handling for diffractive imaging of biological cells, organelles and tissues
- Applications in nanoscale semiconductor devices, strain engineering
- Applications to catalysis and domain formation in complex oxide systems

The X-ray techniques we would like to see implemented at the Coherent Diffraction Imaging beamline are:
i) Lensless imaging of materials using the hard X-ray diffraction patterns surrounding the Bragg peaks to obtain 3D maps of the shapes of the single-crystalline domains and the strains within them. Phasing is achieved by oversampling the continuous diffraction.
ii) Lensless imaging in the forward direction of biological and radiation sensitive samples using cryo freezing and manipulation. The primary advantage over lens-based imaging is that close to 100% quantum efficiency can be realized. Softer X-rays with higher coherence and larger cross-sections may be useful.
iii) X-ray ptychography by scanning a well-defined probe across overlapping regions of an extended sample to provide the phase information for imaging.

CXD for Strain Imaging

The basic methodology for the Coherent X-ray Diffraction (CXD) experiment is to place a spatially coherent beam of X-rays on the sample, so that scattering from all its extremities can be expected to interfere in the diffraction pattern. First suggested by Sayre in 1953 [1], the general method was first demonstrated by Miao in 1999 [2]. The crystal lattice introduces a powerful new constraint on the selection of a grain for imaging. A polycrystalline sample will have closely-packed grains with numerous different orientations. Its Bragg diffraction will resemble that of a powder but, with a small enough beam and typical grain sizes around a micron, the individual grains can still be separated. Even highly textured samples can still have enough distribution of orientations that the grains can usually be distinguished. Once a Bragg peak is isolated and aligned, its internal intensity distribution can be recorded by means of an area detector at the end of a long detector arm. A rocking series of images passing through the center of the Bragg peak yields 3D data, as shown in Fig 1, consisting of characteristic rings resembling the Airy pattern of a compact solid object and streaks attributed to its prominent facets.

For an ideal crystal, meaning its unit cells lie on a perfect 3D mathematical lattice, this distribution is the same around every Bragg peak and indeed about the origin of reciprocal space. Overall inversion symmetry of the diffraction (Friedel's law) also implies the diffraction should be locally symmetric about the center of each Bragg reflection, resulting in symmetric intensity patterns in the CCD. This is sometimes, but not always, observed. When a non-symmetric pattern is seen, it can be decomposed into symmetric and antisymmetric parts. To a good approximation, the symmetric part can be considered to come from the real part of the electron density, while the antisymmetric part is associated with an imaginary density that might represent a component of the strain field projected onto the Bragg peak in use. Once the diffraction pattern is phased, it can be inverted by means of a Fourier transform into a complex density function with the real and imaginary parts interpreted as physical density and strain.

The phasing of the data is a critical step that uses a computer algorithm that takes advantage of internal redundancies when the measurement points are spaced close enough together to meet the "oversampling" requirement. The first step is to postulate a 3D "support" volume in which all the sample density will be constrained to exist. The best method so far for finding those phases and avoiding "stagnation" problems is Fienup's Hybrid Input-Output (HIO) method [3], which starts with a random phase "seed". We now consider our phasing algorithms to be a trustworthy 'black box' tool for data evaluation, so can now start to concentrate on the synthesis of the nanocrystal particles themselves.

We illustrate the capabilities of our CXD method with a recent example of a Pb nanocrystal which was published in Nature in 2006 [4]. The physical density of the crystal was almost constant with no defects, but there was a prominent imaginary part which is attributed to an internal strain field. The figure shows the strain field as an isosurface, a 3D rendering of a single phase contour. The measured strain component is apparently caused by the distribution of contact forces with the substrate upon which the crystal is grown. However, its propagation into the interior must obey the laws of elasticity in a defect-free isotropic medium, as indeed it does. The maximum strain component seen is a phase shift of the complex density of 1.4 radians, corresponding to a total displacement (relative to the ideal crystal lattice) of about a quarter of a Pb{111} atomic spacing, or 0.08 nm.

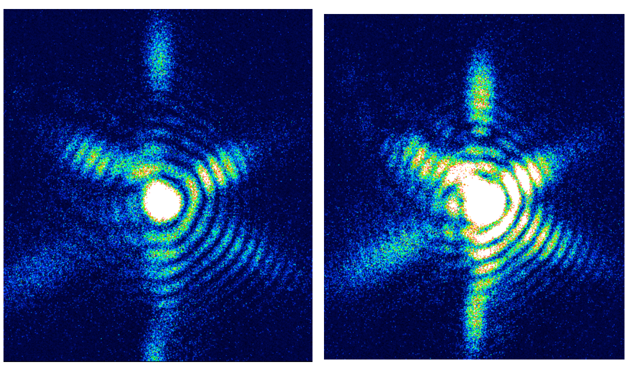


Figure 1. CCD images of X-ray diffraction patterns of a gold nanocrystal, rocking a small angle (<0.1º) near its (111) Bragg peak.

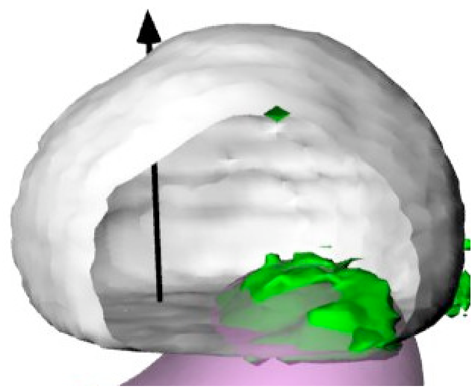


Figure 2. Cut-away isosurface of the density of the Pb nanocrystal obtained with CXD. Inside are superimposed the measured and fitted phase distributions, also at a single contour level [4].

The strain effects in nanocrystals and the deviations from 'bulk' behavior will be more pronounced for smaller crystals. It is therefore essential to focus the beam to reach the smallest possible size range. This is possible using Kirkpatrick-Baez (KB) mirror optics or Fresnel Zone Plates without spoiling the coherence. Our successful inversion of a 160nm cube of silver indicates that the coherence has been sufficiently well preserved by the focusing employed. Once the coherence is defined by an aperture, suitable optics can be introduced to match a wide range of samples while maintaining the coherent quality of the beam.

X-ray Ptychography for Domain Structures

While CXD data from compact, isolated and stable objects can usually be inverted using known methods, it is found to work much less well for continuous objects in the place of isolated nanocrystals because:
i) the illumination profile of the beam on the sample is not well known. The illumination can be defined with an aperture, but apertures with a well defined shape are difficult to make for hard X-rays. The propagation from the aperture, even if placed just millimeters in front of the sample, leads to Fresnel

diffraction and a complex probe beam with strong amplitude and phase variations; this illumination function must be known accurately to define the support, and hence the final structure sought.
ii) there is a high degree of internal symmetry in a typical continuous object, often with multiple copies of separated objects with differing orientations. For example, antiphase domains (APDs) tend to look similar is size and shape. Such symmetry is very bad for unique phasing because interchange of two similar objects within the field of view leads to almost the same overall diffraction pattern; more than two objects would have many possible permutations, all hard to distinguish.

An exciting new direction that gets around this limitation of CXD is the ability to phase diffraction patterns using redundancies introduced by overlapping regions on the sample. Once phased, the diffraction can be Fourier transformed immediately to obtain an image of the sample. X-ray Ptychography is a potentially important new approach to this problem, which was demonstrated for the first time last year [5]. The method involves scanning a small coherent probe over the sample and measuring the diffraction with enough resolution to record all the speckles. Overlapping regions will introduce subtle correlations between the patterns that can be used to extract the missing phases of the diffraction.

Antiphase domain (APD) structures in Cu_3Au were the first samples ever to be investigated by CXD, even before 3rd generation SR sources were available [6]. Strong speckle arises because the submicron domains scatter exactly out of phase with each other. Despite attempts by Eric Dufresne, Mark Sutton and others [7-9], no-one has yet been able to invert such a pattern to obtain an image of the domains. The closest anyone has come to date is the work of Lorenz Stadler's PhD in Vienna, on APD's in an $Fe_{65}Al_{35}$ alloy [10,11], shown in Fig 3. The amplitude of the image is a smooth Gaussian function corresponding to the beam shape, but the colored features are the APDs, shown using a color triangle representing the range $-\pi<\varphi<\pi$. The boundaries are seen to have phase jumps close to π, as they should, but the solution was not unique.

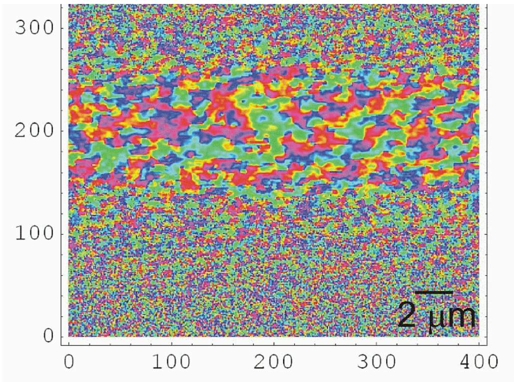


Figure 3. Inversion of an antiphase domain structure in $Fe_{65}Al_{35}$. The elliptical area is the footprint of the X-ray beam on the sample and used as a support for phasing [11].

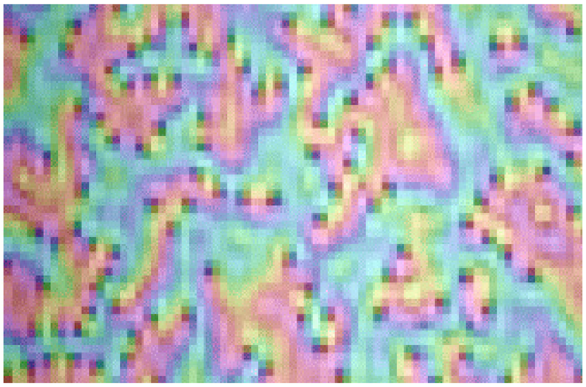


Figure 4. Early attempt at phasing of APD structure in Cu_3Au [7], with phase represented on a color triangle.

There are many outstanding problems with phasing such structures. The phasing algorithms tend to 'stagnate' and introduce 'vortices' into the real-space images. An example of this for Cu3Au is shown in Fig 4, generated during the thesis work of John Pitney [7]. The black dots where the image amplitude goes to zero are surrounded by 2π phase wraps, as seen. The best algorithm for removing them is Fienup's HIO algorithm [3], but it is still not really known why this is so effective.

Ptychography was proposed for electron diffraction by John Rodenburg several years ago, but was found to be difficult to implement. In 2007, he got it working with Franz Pfeiffer using X-rays at the SLS to obtain the image of a zone plate, shown in Fig 5 [5]. The redundancies in the 289 overlapping diffraction patterns allowed inversion of diffraction from a self-similar object.

It has been reported that both the ptychography and HIO algorithms converge faster when a curved wavefront is used. A coherent beam would be focused with a zone plate (or other optics) before scanning across the sample to collect the diffraction. The HIO curved-beam methods, under development by Nugent, Williams, and Peele [22] can be developed much further with the higher coherent flux of NSLS-II. In particular, the “keyhole” method takes advantage of the Quiney solution [26] to measure the complex illumination profile of a curved beam, allowing a sample sub-region to be imaged with a single view [27]. As with ptychography, this new method is promising for imaging non-isolated, extended samples with an arbitrary effective field of view.

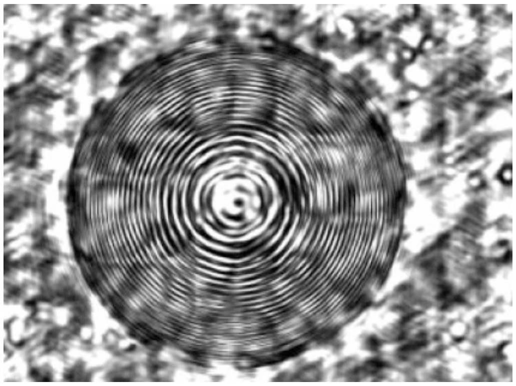


Figure 5. First demonstration of X-ray ptychography: image of a zone plate at the Swiss Light Source [5].

Cryo CXD Imaging of Cells and Organelles

Important results have been obtained in lensless CXD imaging of whole cells using cryogenic sample handling methods. Impressive data shown in Fig 6 have been obtained for the human chromosome by Nishino et al [21]. The image of the yeast cell in Fig 7 was obtained by Shapiro et al [23]. More robust handling of fragile cryo specimens needs low mass, which is appropriate for both specimen scanning and high precision rotation stages. Because of the overlap with CryoEM, it would be good to encourage commercial support, since these are complex systems that benefit from generations of development. NSLS-II will need to develop a new cryo system for the CDI beamline, possibly based on the cartridge design shown in Fig 8. We have expertise at BNL on the BAT in the area of cryo handling.

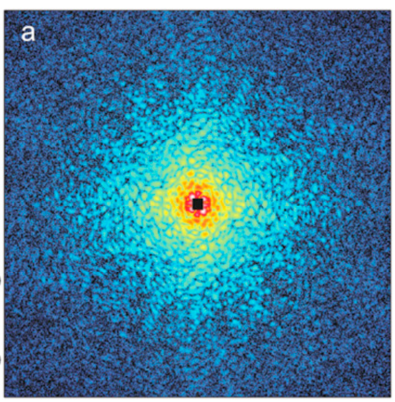


Figure 6. Coherent X-ray diffraction pattern of a HeLa cell chromosome.

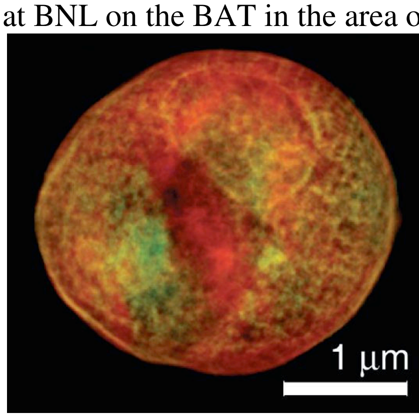


Figure 7. Diffraction reconstruction of a yeast cell taken at 750 eV [23]. Absorption as brightness, phase as hue.

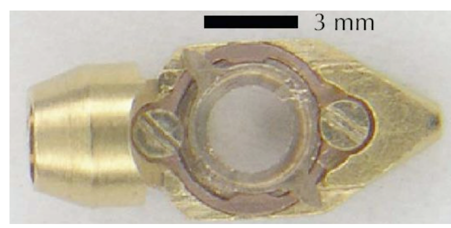
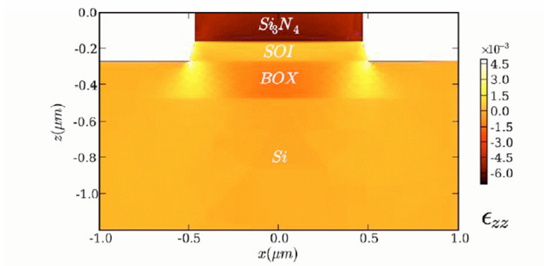


Figure 8. Gatan Martinsried FEI cryo cartridge now used by FEI Polara [24]

Strain Engineering in Semiconductor Devices

Recent indications of the power of CDI in measuring strain in artificial structures has come from the group of Olivier Thomas at the CNRS TECSEN lab in Marseille. They have used high-resolution X-ray diffraction to examine the strain distributions in lithographically prepared structures, both arrays [14] and singular structures [15]. A good example a patterned structure engraved in a Silicon on Insulator (SOI) layer lying on its buried oxide (BOX) substrate and underlying bulk Si handle, as illustrated in Fig 9. An array of 1μm wires was cut in the SOI (100nm Si on 200nm SiO_2) using a Silicon Nitride (SiN) lithography mask. Significant distortions in the Si wire, calculated using Finite Element Analysis (FEA), are shown. The kinematical diffraction pattern of the strained wire, shown at the bottom of Fig 9, is in good agreement with the experiment.

Averaging is used to increase the signal in the experiment, which was carried out at BM32, a bending magnet beamline at ESRF. Slight disorder in the relative positions of the wires making up the array removes any effect of interference between them; the limited coherence of the BM32 beam, in the range of several microns, would not be enough to achieve this. But since the wires are effectively floating on amorphous oxide, it is reasonable that any spatial correlations present in the parent SOI layer would be lost. Future version of this experiment, as proposed for the CXD beamline at NSLS-II, using focused undulator radiation would have no trouble seeing individual wire structures. Objects as small as individual InP semiconductor nanowires far below 100nm diameter, containing at least ten times less material than a single one of the SOI structures of Gailhanou et al [15], have been measured at 34-ID-C and did not need to be built in arrays.



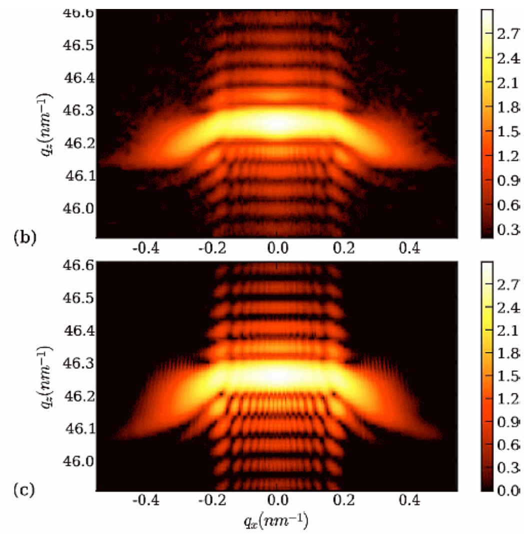


Figure 9. Above: FEA calculation of the strain distribution in a strip of patterned Silicon on Insulator (SOI). The supporting oxide layer is denoted BOX. Right: measured and calculated diffraction patterns [15].

These samples have large strains with many ‘phase wraps’ present in the structure. Indeed, without strain, the SOI wires would give diffraction patterns extending less than one twentieth of the width seen in Fig 10. The X-ray methods discussed here are sensitive to stresses orders of magnitude smaller than this. Yet the fabrication method of Gailhanou et al [15] is common for constructing devices with the 45nm design rules in production today.

The CDI beamline at NSLS-II can go beyond the achievements of Thomas et al by looking at individual strained structures cut into SOI. Strain patterns can be created in model devices with sizes more relevant to current technology (45nm), that penetrate partially into the thickness of the SOI layer, as is relevant. As Thomas et al found, CXD is particularly valuable in devices fabricated using SOI because the active layer of Si has a different orientation from the much thicker handle; the diffraction of interest would be in the shape of the 111 or 220 Bragg peak of the layer, which would be swamped by the bulk diffraction if SOI technology were not employed.

Complex Oxides, Phase Structures, in-situ Chemical Reactions and Catalysis

Coherence techniques can be applied to the dynamics of complex oxide materials driven far from equilibrium by external electric fields. X-ray scattering techniques couple directly to the order parameters relevant to ferroelectricity and magnetism, and are fully quantitative [25]. Coherent scattering can resolve small strains with spatial resolution far better than selected-area diffraction approaches. In addition, coherent scattering can have time resolutions limited only by the bunch length of the pulses from the storage ring, which will be on the order of 15 ps at NSLS-II. Phenomena accessible with the dramatic advance in spatial and temporal resolution include domain dynamics, the physics of magneto-electric coupling, the coupling of soft modes to applied fields, coupling of strain between components of multilayers and multicomponent multifunc-tional materials. Coherent scattering from magnetic order (resonant or non-resonant) has the potential to probe magnetic dynamics in buried systems. Time-resolved coherence techniques can be extended to the dynamics of electronic and magnetic systems driven far from equilibrium with applied electric fields.

Imaging methods will be applied to phase structures in electronically and magnetically ordered systems showing Charge, Orbital, and Spin ordering, such as the domain structure of the technologically important CMR manganites. The organization of charge and orbital domains within these strongly correlated electron systems has interesting dynamic behavior near phase transitions [28]. Unlike micro/nano diffraction, coherent imaging is sensitive to the relative phases, rather than orientation of order parameter. The CXD beamline could answer why there is substantial inhomogeneously distributed “strain” of the CDW/SDW wavevector in Cr, for example. This strain varies very slowly following a temperature change, in a fashion similar to ageing of glassy materials. It is important to find out if there is a relationship between these “electronic crystal” defects and the strain in the lattice.

Imaging of dislocations and other phase defects in crystalline materials will be another important direction. For example, one could directly track dynamics of defects as the temperature is changed. Other examples are the study of dynamics of antiphase domain walls in ordered binary alloys, premelting and phase transitions in nanomaterials. One could track the formation of non-crystalline “quasi-liquid” layer by watching the surface areas of a particle or nanowire disappear. Lastly, there are “Nanoconfined” materials with nanosized crystalline inclusions in some other matrix. Using CXD, the enclosing matrix is basically invisible, and only the inclusions are visible. These deeply buried systems present a unique opportunity where electron microscopy is completely inappropriate.

B. Beamline Concept & Feasibility

- Canted low-beta IVU20 undulators with monochromatic beams 2.5-20keV(A) and 8-20keV(B)
- Long beamlines (200m) for maximum demagnification
- Branch A for biological in-line CDI; branch B for Bragg diffraction CDI.
- Ultra-stable optical bench in mechanically isolated external building
- Ultra-high precision position stage for sample and optics in controlled environment
- Cryo sample stage with single rotation axis and in-vacuum detector (A)
- Full angle range (3 degrees of freedom) for access to reciprocal space points (B)
- Quantum efficient area detectors, mechanically isolated from sample with high dynamic range

In both the original planning for NSLS-II, and all previous coherent beamline designs, it has been safe to assume that the coherence lengths are so much smaller than the beam cross section that multiple branches can be fed in tandem. This has been achieved successfully at 8-ID and 34-ID of APS. It has come to light recently [18] that the coherence of the NSLS-II design is so high that this will no longer be possible, even in the horizontal direction. This is not because the beam is fully coherent in that direction, but because the practical location (shield wall) where the beam can be split is so far away that a secondary source (i.e. horizontal slit in the front end) placed there would not be filled with light to work correctly [18]. The secondary source idea is attractive because it decouples the stability of the storage ring from the beamline, so can potentially produce a more stable beam. The possibility of a refocussed secondary source using an optics hutch inside the synchrotron building will be considered as an additional option (see below).

Low-beta straights at NSLS-II are significantly brighter than the high-beta ones (and even less suitable for making a secondary source with a slit [18]) and also more numerous on the floor plan. At the present time, the IVU20 high-field (cryogenic, permanent magnet) in-vacuum undulator design appears to be the best choice. The undulators should be made as long as possible with a period of about 18-20mm (depending on the minimum gap allowed). They should tune over most of the range 2.5-20keV, but could allow some gaps if there are significant gains at certain other energies.

The long beamline is needed for the same reasons as at Diamond, Soleil, eventually at ESRF and potentially at APS to give the maximum combination of demagnification and working distance around the sample. It is expected that high-demagnification (eg 1000×) optics will be used in front of the sample, probably Kirkpatrick-Baez (KB) mirrors to match the coherent beam size to that of the nano-scale object being studied. Space around the sample is needed for sample environment and for multiple parallel probes, microscopes, alignment tools etc. It is estimated that 200m would be a good length of the beamline, taking it well outside the main building in a design similar to Spring8 (250m and 1km), ESRF (150m) and Diamond (250m). Christoph Rau (Diamond) has found that building the entire structure, including hutches, from concrete leads to significant cost savings and that a long beamline does not cost much more.

Beamline optics will be minimal in order not to disturb the coherence. Double crystal monochromators, (horizontally deflecting for branch B) will be located just in from of the sample hutch. Si(111) crystals will be able to reach (at 52°) the 2.5keV lowest energy specification of branch A. By spreading the heat load, this can be water-cooled, hence inexpensive and more stable. Horizontal harmonic filter mirrors, are needed in the Front End Optics (FOE) hutch, as close to the source as possible so that any imperfections can be considered part of the source. This has the safety advantage of removing all Bremstrahlung before the beam leaves the storage-ring building.

The FOE, located adjacent to the storage-ring shield wall, will be the only hutch of the beamline in the storage-ring building. It will be made large enough for the introduction of additional optics for upgrading the beamline. Use of a vertical collimator in the FOE would contain the full coherent flux within a narrower fan and hence reduce the size of the focusing optics needed just before the sample. The collimator could be a selection of compound refractive lenses (CRLs) for the different wavelengths, kinoform lenses, or else curved mirrors. It is not clear at present whether such a white-beam collimator can be made sufficiently accurately, however. To the extent that this element can be considered as part of the source, its imperfections should only spill flux and not adversely affect the coherence function. Investment in an optics R&D program on the part of NSLS-II would be highly valuable to help evaluate these options.

Apart from a possible secondary-source slit in the FOE, the only slits will be for defining the coherence just in front of the focusing optics and sample. In the case of ptychography, the optics may be replaced by a pinhole or some other condenser system. The focusing system should be in vacuum to keep it clean and the vacuum may also enclose the sample and its environment to avoid using windows. On the soft X-ray branch (A) going down to 2.5 keV, windowless operation is highly desirable. On each branch, the optics and sample will be on a common high-stability optical table to minimize their relative vibrations. A good plan for this is to isolate the table completely from the building by placing it on its own piles into the ground as done at Diamond. Both sample and optics will be on ultra-high precision stages with encoders, possibly interferometers. For diffraction, at least one rotation axis will be needed, but 3 degrees of freedom would be better. It should be noted that there has been significant progress in the engineering of nanopositioning systems in the past few years. There has been a revolution in the design of such instrumentation, from which NSLS-II can benefit immediately.

The revolutionary aspect of the design is to keep the mechanical parts of the detector system as far away from the sample as possible, with as little physical connection as possible. Heavy detector systems on long arms are totally incompatible with the nm-level precision of the sample and optics. The detector only has to be as stable as its pixel size, or a fraction of a millimeter, which can be achieved by free standing robots. A detector distance range from 1–4m is envisaged for the robot on branch B. If funds are available, a stationary large pixel array covering a large solid angle might be an option. The in-vacuum detector on branch A will need to be 10m away because the field-of-view of the sample is larger. Unless a very high counting rate detector can be developed, it will need a carefully designed beam stop.

A serious limitation in the past has been the dynamic range of the available detectors, often below 10^2 for CCDs. A number of new detectors is becoming available, such as Pilatus, MediPix, and CMOS Monolithic Active Pixel Sensor (MAPS) technology. The trend is towards larger pixels with circuitry, such as thresholding, gain control or counting, included on each pixel. As always, it is hoped that NSLS-II will invest significantly in developing detectors. The angular pixel size needed for the CXD is determined by the particle size and tends to be quite large, allowing for closer distances. For the forward-scattering CDI, and usually for ptychography, smaller pixels and bigger distances would be needed because the beam size on the sample is set by a pinhole, perhaps 10μm in diameter, which would need 100μm pixels, 10m away.

Sample environments would need to fit in the confined space between the focusing optics and the sample goniometer of branch B. Use of a long beamline ensures the maximum clearance for the application of electric and magnetic fields, hot and cold temperatures, high pressure cells, vacuum chambers and space for user-supplied instrumentation. The soft X-ray instrument on branch A will need careful vacuum design to avoid placing windows where they can affect the coherence. The cryo-sample manipulator will be designed to allow rapid sample introduction and exchange, with sufficient stability and tilt degrees of freedom for 3D lensless imaging. A cryo sample prep lab with plunge freezing capabilities, perhaps employing some of the expertise from Baumeister's group in Martinsried, could be shared with the STXM beamline of NSLS-II and with the CryoEM facilities located elsewhere at BNL. Prescreening by optical microscopy would help with controlling of the ice quality in the samples before they are inserted.

C. Required Technical Advances

i) It should be considered whether we can employ the 'mini-beta' concept, which has allowed utilization of long straight sections for canted undulators at both Soleil and Diamond. Extra quadrupole magnets in the machine lattice allows conversion of a less useful high-beta straight into two low-beta sources.
ii) Concrete shielding, instead of lead, could be used to save money on the external part of the construction. With a long beamline there is plenty of separation of the canted branches to allow this. A major advantage of the external building is that it will be mechanically isolated from the storage ring building. The ground stability and effect of disturbance by personnel could be mitigated by the use of a separate control room.
iii) High quality detectors covering a wide solid angle will be needed to extract the most information from the experiments. There may be some synergy with other beamlines in the second phase of NSLS-II, who could share the development costs, remembering that a long time-scale is needed to develop new detectors.
iv) Secondary source options within the synchrotron building should be explored.

D. User Community and Demands

As mentioned by Ray Orbach when he approved the project, Coherent X-ray Diffraction Imaging was one of the original justifications for building NSLS-II. It was argued, based on various published calculations [19], that the >10× increase of brightness of NSLS-II over all previous sources would allow the methods to reach atomic resolution. Subsequent discussion has indicated there a radiation damage limit may preclude this possibility even for 'hard' materials [20], but other advances have intervened:
i) the realization of the importance of phase contrast imaging for studying both structure and strain, where atomic resolution is less relevant (or meaningful) [4]
ii) the potential of the ptychography and keyhole methods [5, 27] for extended samples
iii) the need for custom-designed cryo-handling of biological samples in vacuum for diffraction imaging and ptychography, in a way that is compatible with STXM and CryoEM instruments at BNL.

Various workshops have been held that demonstrate the depth of the potential user community. In all cases the recommendations have been to accelerate the development of the CXDI techniques. While this is far from being a complete list, it does appear that the main growth of the field has been in Europe and that NSLS-II will be strategically placed to bring some of those advances back to the USA:

- Diamond Light Source Workshop on Coherence, March 2004
- Workshop on emerging new directions of synchrotron research, SRC, Madison, September 2004
- Journée Scientifique sur Diffraction et Diffusion des X Coherents, CNRS Grenoble, October 2004
- BESSY Coherence Workshop, Berlin, December 2004
- Phase Retrieval and Coherent Scattering Workshop, Isle Porquerolles, France, June 2005
- Workshop on Diffraction, Crystallography and Imaging at the XFEL, Hamburg, October 2005
- ERL workshop, Cornell University, New York, June 2006
- Coherence/Imaging Planning Meeting, Advanced Photon Source, July 2006
- Symposium on Nanoscience, Diamond Light Source Users Meeting, September 2006
- SLS-Soleil Workshop on The Full Spectrum, Villigen, September 2006.
- Workshop on coherent x-ray microscopy (APS Users Meeting), 8-9 May 2007
- ESRF Science at the Nanometre Scale Workshop, October 2007
- Workshop on Coherent X-ray Diffraction at NSLS II, Brookhaven, March 2008
- Heraeus Seminar Matter in Coherent Light, Bad Honnef, Germany, March 2008
- X-ray Coherence Workshop, Stanford Linear Accererator Center, Stanford, October 2008
- ID01 Upgrade Workshop, European Synchrotron Radiation Facility, Grenoble, France, December 2008
- 20th International Congress on X-ray Optics and Microanalysis, Karlsruhe, Germany, September 2009
- Laser Science XXV, 25th Annual meeting of APS/DLS, San Jose, October 2009
- X-ray coherent diffraction Workshop & XFEL Meeting, Soleil, France, December 2009
- Scientific Potential of Free Electron Lasers, The Royal Society, London, April 2010
- XFEL workshop, Zernike Institute for Advanced Materials, Groningen June 2010
- Coherence 2010, Rostock, Germany, June 2010

E. Proposal Team Expertise and Experience

The proposed BAT members are the authors of this proposal. Ian Robinson (University College London, spokesperson) has developed the CXD method with the construction of beamline 34-ID at APS. Oleg Shpyrko (University of California, San Diego) has been a major user at NSLS and of CXD at APS. He is building a university-based group of future users with interests in nanoscale pattern formation in complex systems. Paul Evans (University of Wisconsin) has developed time-resolved experiments at APS applied to ferroelectrics and multiferroics. Chris Jacobsen (Argonne National Lab) participated in the development of the X1 undulator for x-ray microscopy, developed the STXM instrument there, and is involved with the COSMIC beamline project for CXD imaging at ALS. Joe Wall (Brookhaven National Lab) leads the STEM biological imaging group and is an expert in cryo-freezing for sample preparation. Christoph Rau (Diamond Light Source) has developed phase-contrast full-field microscopy at APS and is currently building the Coherence and Imaging beamline at Diamond. Paul Fuoss (Argonne National Lab) has built surface diffraction beamlines at NSLS and SSRL and is involved in planning single molecule diffraction experiments at LCLS and APS. Ian McNulty (Argonne National Lab) has developed x-ray microscopy, nanofocusing, and coherent diffraction methods for the intermediate-energy region. He previously led the X-ray Microscopy Group based at APS sector 2 and is planning a new coherent diffraction initiative at APS.

F. Suggestions for BAT Membership

Ian Robinson (spokesperson), University College, Gower St, London, WC1E 6BT, i.robinson@ucl.ac.uk

Oleg Shpyrko, University of California at San Diego, La Jolla, CA 92093, oshpyrko@physics.ucsd.edu

Paul Evans, University of Wisconsin, 1509 University Avenue, Madison, WI 53706, evans@engr.wisc.edu

Chris Jacobsen, Argonne National Lab, Argonne IL 60439, chris.j.jacobsen@gmail.com

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G. References

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Ian Keith Robinson

Education

1976-81	Harvard University, Cambridge, Massachusetts. Ph.D. Biophysics. Thesis "The structure of the expanded state of tomato bushy stunt virus". Advisor: Prof. Stephen Harrison.
1973-76	Cambridge University, England. M.A. Natural Sciences. First Class Honours. Thesis "A study of a position sensitive neutron detector".

Employment

1981-92	Distinguished member of technical staff, AT&T Bell Laboratories. Basic research in the Chemical Physics Laboratory.
1990-91	Professeur (Chaire Municipale), Université de Grenoble
1992-2005	Professor of Physics, University of Illinois at Urbana-Champaign
2006-pres	Professor of Physics and Astronomy, University College, London
2006-pres	Diamond Fellow, Diamond Light Source, Oxford

Honors

Fellow of the American Physical Society
Bell Labs distinguished member of technical staff (DMTS)
Chair of X-ray Physics Gordon Conference, 1995
Co-Chair of X-ray Physics Gordon Conference, 1993
Co-Chair of Surface X-ray and Neutron Scattering conference (SXNS-2), 1991
Editor, Journal of Physics: Condensed Matter, 1991-6
Organizer of NSLS Workshop on "In-situ Manipulation", 1998
B. E. Warren Award winner of American Crystallographic Association, 2000
Chair of IUCr Synchrotron Radiation Commission, 2002-5
Center for Advanced Studies Associate, University of Illinois, Urbana, 2003-4
Ted Maslen Award of the Society of Crystallographers in Australia and New Zealand, 2003
Organizer of Euro-XFEL Workshop on “Diffraction, Crystallography and Imaging”, 2005
Humboldt Foundation, Senior Research Fellowship, 2004-6
Organiser of LCN MedX workshop on Medical Imaging, 2006

Synchrotron-related Committees

Advanced Photon Source Users Organization Steering Committee, 1992-5
National Synchrotron Light Source Proposal Study Panel, 1994-6
National Academy of Sciences National Committee on Crystallography, 1994-7
National Synchrotron Light Source User Executive Committee, 1996-8
UNICAT (Argonne National Laboratory) Board of Governors 1997-2005
National Academy of Sciences National Committee on Crystallography, 1999-2001
International Union of Crystallography Synchrotron Radiation Commission, 1999-2005
Surfaces and Interfaces Review Committee, European Synchrotron Radiation Facility, 2001-2004
Experimental Facilities Advisory Committee, Spallation Neutron Source, 2002-2004
Science Advisory Committee, Diamond Light Source, 2004-2006
Science Advisory Committee, Soleil Light Source, 2006-pres
National Synchrotron Light Source Experimental Facilities Advisory Committee, 2006-9

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Education and Training

1994, B.S., *Engineering Physics*, Cornell University
1996, S.M., *Applied Physics*, Harvard University
2000, PhD, *Applied Physics*, Harvard University
2000-2002, Postdoctoral Research, *Physical Sciences Research Division,* Bell Labs, Lucent Technologies

Research and Professional Experience

September 2002-2008, Assistant Professor, Materials Science and Engineering, University of Wisconsin-Madison

September 2008-present, Associate Professor, Materials Science and Engineering, University of Wisconsin-Madison

Selected Publications

"*Piezoelectricity in the Dielectric Component of Nanoscale Dielectric-Ferroelectric Superlattices*," J. Y. Jo, R. J. Sichel, H. N. Lee, S. M. Nakhmanson, E. M. Dufresne, and P. G. Evans, Phys. Rev. Lett. **104**, 207601 (2010).

"*Stability of the unswitched polarization state of ultrathin epitaxial Pb(Zr, Ti)O_3 in large electric fields*," A. Grigoriev, R. J. Sichel, J. Y. Jo, S. Choudhury, L.-Q. Chen, H. N. Lee, E. C. Landahl, B. W. Adams, E. M. Dufresne, and P. G. Evans, Phys. Rev. B **80**, 014110 (2009).

"*Nonlinear piezoelectricity in epitaxial ferroelectrics at high electric fields*," A. Grigoriev, R. Sichel, H. N. Lee, E. C. Landahl, B. W. Adams, E. M. Dufresne and P. G. Evans, Phys. Rev. Lett. **100**, 027604 (2008).

"*Synchronizing fast electrically driven phenomena with synchrotron x-ray probes*," A. Grigoriev, D. Dal-Hyun, P. G. Evans, B. Adams, E. Landahl and E. M. Dufresne, Rev. Sci. Instrum. **78**, 23105-23101 (2007).

"*In Situ X-Ray Probes for Piezoelectricity in Epitaxial Ferroelectric Capacitors*," D.-H. Do, A. Grigoriev, D. M. Kim, C.-B. Eom, P. G. Evans, and E. M. Dufresne, Integrated Ferroelectrics **101**, 174 (2009).

"*Nanosecond Domain Wall Dynamics in Ferroelectric Pb(Zr,Ti)O_3 Thin Films*," A. Grigoriev, D.-H. Do, D. M. Kim, C.-B. Eom, B. Adams, E. M. Dufresne, and P. G. Evans, Phys. Rev. Lett. **96**, 187601 (2006).

Synergistic Activities

Advanced Photon Source Condensed Matter Physics general user proposal review panel (2003-2005), Argonne Center for Nanoscale Materials User Executive Committee (2005-2009, chair 2006-2008), Advanced Photon Source User Organization Executive Committee (2006-2009), Special Interest Group Representative to National Synchrotron Light Source User Executive Committee (2006-2007). Member of American Physical Society, Materials Research Society, AAAS, and Tau Beta Pi Honor Society. University of Wisconsin Polygon Engineering Student Council Outstanding Instructor Award (2006, 2007).

Chris Johnson Jacobsen, Northwestern University/Argonne Lab

Education:

St. Olaf College, Northfield, Minnesota: B.A. (Physics, *cum laude*), 1983
State University of New York at Stony Brook: PhD (Physics), 1988

Employment:

2010-present: Associate Division Director, X-ray Science Division, Advanced Photon Source, Argonne National Laboratory
2010-present: Professor, Dept. Physics & Astronomy, Northwestern University
1989-2010: Dept. Physics & Astronomy, Stony Brook University (Postdoc 1989; Asssistant Professor 1991; Associate Professor 1995; Professor 2000)
8/2007-1/2008: Chief Scientist, Xradia Inc., Concord, CA (while on leave from Stony Brook)
1988-1989: Postdoc, Center for X-ray Optics, Lawrence Berkeley National Laboratory
1980-1982: summer undergraduate student, Los Alamos National Laboratory

Honors, Awards:

Presidential Faculty Fellow Award (White House/National Science Foundation), 1992—1997.
International Dennis Gabor Award (Hungary, for work in modern optics before age 35), 1996.
Fellow: Optical Society of America, 1999; American Association for the Advancement of Science, 2002
R&D 100 award (R&D Magazine: 100 most technologically significant new products and processes of the year): Cryo STXM I (Maser *et al.*), 1999
Kurt Heinrich Award (outstanding young scientist), Microbeam Analysis Society, 2001.
Most Outstanding Teacher award (Dept. Physics & Astronomy, Stony Brook University): 1996, 2002.

Other Activities:

Editorial board, *Ultramicroscopy*, 1995-present
Lecturer at the Hercules (High European Research Course for Users of Large Experimenal Systems) courses on Biomolecular Structure and Dynamics, 2004-present
FASR Distinguished Lecturer, Cornell Physics/CHESS (2006), DAAD Fellowship (Germany, 1995), Institute of Applied Study Distinguished Visiting Fellow, LaTrobe University (Australia, 2006)
Scientific Advisory Committee, Advanced Light Source (2009-present), Diamond/UK (2010-present)

Invited Presentations:

90 in North America, 60 in Europe, 30 in Asia.

Recent publications:

1. J. Nelson, X. Huang, J. Steinbrener, D. Shapiro, J. Kirz, S. Marchesini, A.M. Neiman, J.J. Turner, and C. Jacobsen, "High resolution x-ray diffraction microscopy of specifically labeled yeast cells," *Proceedings of the National Academy of Sciences* **107**, 7235 (2010).
2. X. Huang, J. Nelson, J. Kirz, E. Lima, S. Marchesini, H. Miao, A.M. Neiman, D. Shapiro, J. Steinbrener, A. Stewart, J.J. Turner, and C. Jacobsen, "Soft x-ray diffraction microscopy of a frozen hydrated yeast cell," *Physical Review Letters* **103**, 198101 (2009).
3. M. D. de Jonge, B. Hornberger, D. Paterson, C. Holzner, D. Legnini, I. McNulty, C. Jacobsen, and S. Vogt, "Quantitative phase imaging with a scanning transmission x-ray microscope," *Physical Review Letters* **100**, 163902 (2008).
4. J. Lehmann, D. Solomon, J. Kinyangi, L. Dathe, S. Wirick, and C. Jacobsen, "Spatial complexity of soil organic matter forms at nanometre scales," *Nature Geoscience* **1**, 238-242 (2008).
5. D. Shapiro, P. Thibault, T. Beetz, V. Elser, M. Howells, C. Jacobsen, J. Kirz, E. Lima, H. Miao, A. M. Neiman, and D. Sayre, "Biological imaging by soft x-ray diffraction microscopy," *Proceedings of the National Academy of Sciences* **102**, 15343-15346 (2005).
6. C.K. Boyce, M.A. Zwieniecki, G.D. Cody, C. Jacobsen, S. Wirick, A.H. Knoll, and N. M. Holbrook, "Evolution of xylem lignification and hydrogel transport regulation." *Proceedings of the National Academy of Sciences* **101**, 17555-17558 (2004).
7. J. Kirz, C. Jacobsen, and M. Howells, "Soft x-ray microscopes and their biological applications." *Quarterly Reviews of Biophysics* **28**, 33-130 (1995).

Biographical Sketch: Oleg G. Shpyrko

Education:

Ph.D., Physics, Harvard University, 6/2004
B.Sc., Physics, Moscow Institute of Physics and Technology, Russia, 6/1998

Employment:

July 2007 – present	Assistant Professor, Department of Physics, University of California San Diego
2005 – 2007	Distinguished Postdoctoral Fellow, Center for Nanoscale Materials, Argonne National Laboratory
2004 – 2005	Postdoctoral Fellow, Division of Engineering and Applied Sciences, Harvard University
1998 – 2004	Graduate Research Assistant, Department of Physics, Harvard University

Honors, Awards:

- NSF CAREER Faculty Award, 2009;
- Hellman Foundation Faculty Award, 2009;
- University of California San Diego Faculty Career Development Program Award, 2009;
- Rosalind Franklin Young Investigator Award, Argonne National Laboratory, Argonne, IL, 2008;
- CNM Distinguished Postdoctoral Fellow, Argonne National Laboratory, Argonne, IL, 2005 – 2007

Selected Professional Activities:

LCLS Users' Organization Executive Committee, Stanford Linear Accelerator Center; XPCS Beam Advisory Team, NSLS-II, Brookhaven National Laboratory; Advanced Photon Source Proposal Review Panel, Argonne National Lab; Advanced Light Source Proposal Review Panel, Lawrence Berkeley National Laboratory; Focus Topic Organizer, APS March Meeting, Portland, Oregon 2010; Lecturer, International Research Course on New X-Ray Sciences, DESY Hamburg, Germany 2010; Lecturer, 2nd International School and Workshop on X-ray Micro and Nanoprobes (XMNP2009), Palinuro, Italy, 2009; Lecturer, School for Liquid Surface X-ray Scattering, Argonne, IL 2007

Selected Recent Publications:

1. O. G. Shpyrko, E. D. Isaacs, J. Logan, Y. Feng, R. Jaramillo, H. C. Kim, T. F. Rosenbaum, G. Aeppli, M. Sprung, S. Narayanan and A. Sandy, “Direct measurement of antiferromagnetic domain fluctuations” *Nature* 447, 68 (2007)
2. O. G. Shpyrko, R. Streitel, V. S. K. Balagurusamy, A. Yu. Grigoriev, M. Deutsch, B. M. Ocko, M. Meron, B. Lin and P. S. Pershan “Surface Freezing in Gold-Silicon liquid alloy”, *Science* 313, 77 (2006)
3. “Local Structural Probes”, O. G. Shpyrko, McGraw Hill 2010 Yearbook of Science & Technology (2010)
4. O. G. Shpyrko, A. Grigoriev, R. Streitel, D. Pontoni, P.S. Pershan, B.M. Ocko, M. Deutsch, “Surface induced atomic scale demixing in BiSn eutectic alloy”, *Phys. Rev. Lett.* 95, 106103 (2005)
5. R. Jaramillo, T. F. Rosenbaum, E. D. Isaacs, O. G. Shpyrko, P. G. Evans, G. Aeppli and Z. Cai, “Microscopic and Macroscopic Signatures of Antiferromagnetic Domain Walls” *Phys. Rev. Lett.* 98, 117206 (2007)
6. Y. Feng, R. Jaramillo, G. Srajer, J. C. Lang, Z. Islam, M. S. Somayazulu, O. G. Shpyrko, J. J. Pluth, H.-k. Mao, E. D. Isaacs, G. Aeppli, T. F. Rosenbaum, “Pressure-Tuned Spin and Charge Ordering in an Itinerant Antiferromagnet” *Phys. Rev. Lett.* 99, 137201 (2007)
7. O. G. Shpyrko, P. Huber, P.S. Pershan, B.M. Ocko, H. Tostmann and M. Deutsch “X-ray Study of the Liquid Potassium Surface: Structure and Capillary Wave Excitations” *Phys. Rev. B* 67, 115405 (2003)
8. K. J. Alvine, O. G. Shpyrko, P. S. Pershan, K. Shin and T. P. Russell, “Capillary filling of anodized alumina nanopore arrays”, *Phys. Rev. Lett*. 97, 175503 (2006)
9. E. DiMasi, H. Tostmann, O. G. Shpyrko, P. Huber, B. M. Ocko, P. S. Pershan, M. Deutsch and L. E. Berman, “Pairing Interactions and Gibbs Adsorption at the Liquid Bi-In Surface”, *Phys. Rev. Lett.* 86, 1538 (2001).
10. H. Tostmann, E. DiMasi, O. G. Shpyrko, P. S. Pershan, B.M. Ocko and M. Deutsch, “Microscopic Structure of the Wetting Film at the Surface of Liquid Ga-Bi Alloys”, *Phys. Rev. Lett.* 84, 4385 (2000)